ESTABLISHMENT OF EQUIPMENT AND ITS OPERATION FOR HYDROTREATING OF A COAL-DERIVED LIQUID

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ESTABLISHMENT OF EQUIPMENT AND ITS OPERATION FOR HYDROTREATING OF A COAL-DERIVED LIQUID

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CHAPTER I

INTRODUCTION

Human activities are carried out with expenditure of energy, and the life of industry depends upon the supply of energy. The rate of energy consumption also indicates the growth and productivity of a nation. Being a highly industrialized nation, the United States' total energy consumption is expected to increase from 45 quadrillion (10^{15}) BTU in 1960 and 68 in 1970, to a projected 100 in 1980 (22). The main sources of energy supply are fossil fuels, hydroelectric power, nuclear power, and solar energy. Though only fossil fuels and hydroelectric power are economically feasible for use on a broad basis, nuclear power plants are also being constructed now. Since hydroelectric power is limited, fossil fuels are the major source of energy supply. Among the fossil fuels, coal, natural gas, and oil are mostly used because of their convenient accessibility. Oil is the dominant fuel in transportation because of its ease in use as a liquid, its high combustion value, and its present low cost. The growing need for oil was indicated by a recent announcement (3) which said, "President Nixon has set the authorized level of oil imports to states east of the Rocky Mountains in 1972 at 1,550,000 bbl a day. This represents an increase of 100,000 bbl a day from the 1971 level for imports of crude oil, unfinished oil and refined products..." As 86 percent of Free World

oil reserves outside the United States are controlled mostly by countries in the Arab bloc, Harold Davis (5) pointed out that this nation's energy outlook for the next 15 years may be summed up as too little supply of natural gas and too much dependence on foreign oil. Such dependence must be removed, along with its threat to the nation's economic health and military security. Hence, the extraction and conversion of coal, tar sand, and oil shale into oil and gas and the use of nuclear power or solar energy as substitutes are under development as alternatives to provide a more secure basis of energy supply. Since the development of nuclear power has been behind schedule owing to unforeseen difficulties in construction, and the utilization of solar energy is at its early research state, the technology of converting coal to oil and gas must be employed to meet the not too distant energy demands. Besides, the United States has an abundance of coal reserves. As more coal is processed to liquids and gases, the need for suitable catalysts to remove undesirable heteroatoms from coal-based liquids is expected to increase.

This is a preliminary study on hydrogenation of a coal-derived liquid with an aim to develop the apparatus and analytical techniques for a more detailed program. The major variables in this work are limited to temperature, pressure, and space time. Apart from the central goal, this study also serves to determine the effects of the variables with respect to desulfurization, denitrogenation, and hydrocracking of a coal-derived liquid. Determination of such effects is essential for establishing a reference set of data necessary for catalyst development comparisons.

A trickle-flow reactor was employed in this study. The catalyst,

Nalcomo 474 (see Table III), was selected because it has been satisfactorily used in the petroleum industry for similar service. Since the nature of this study is quite exploratory, the boundaries of the variables were not decided at the beginning of this work; rather, they were to be established as part of the objective. At the start, the first set of values for the variables was estimated from literature. Most reports of related studies indicate that temperature range goes from 662°F (350°C) to 950°F (510°C), and pressure ranges up to 9000 psig. Comparison of space times used by previous research workers is difficult, as space time has a liberal basis of definition. Unless both the density of the feed oil and that of the catalyst are specified, the units of a space time cannot be converted from volume basis to weight basis, or vice versa. Thus, the selection of space times was somewhat arbitrary.

CHAPTER II

LITERATURE REVIEW

The history and technology of hydrogenation of coal and coal liquids have been thoroughly described by Wu and Storch (25) for the available literature through 1967. Owing to shortage of petroleum reserves and plenty of coal reserves, reliance upon conversion of coal to liquid fuel to meet the present and future need has been pointed out by Mills (13). The intention of this chapter is to present pertinent parts of recent studies on hydrogenation of coal liquids in order to serve as a basis for development of and comparison with the present work.

The experimental systems for hydrogenation of coal liquids are distinguished partly by the kinds of reactor which they contain. Reactors are usually built for continuous or batch processing; thus, experimental systems can be classified essentially into those for continuous processing (8, 9, 14, 24) and those for batch processing (4, 15).

Reports of several previous studies show agreement in that desulfurization and denitrogenation increase with rising temperature (2, 14, 15). In hydrocracking of a low temperature coal tar, Qader and Hill (15) employed a batch process with a catalyst consisting of sulfides of tungsten and nickel supported on silica-alumina. This low temperature coal tar was prepared from bituminous coal by carbonization at 1022°F

(550°C). It had an initial boiling point of 192°F (89°C), and a pitch point of 670°F (360°C). In their work, 100 percent removal of sulfur and nitrogen was achieved at 842°F (450°C) for six hours, and 932°F (500°C) for five hours, respectively, and the pressure was kept constant at 1500 psig. They also studied the influence of pressure and found that desulfurization and denitrogenation rates increase with increasing pressure in the range of 1000 psig to 3000 psig. Anderson, et al. (14) studied heteroatom removal by hydrogenation of COED oil. This oil is characterized by its boiling range from 176°F (80°C) to 810°F (432°C). With a trickle-flow reactor containing nickel-molybdenum catalyst, they operated at a constant pressure of 3000 psig, at temperatures ranging from 640°F (338°C) to 790°F (421°C), and at space velocities ranging from 0.8 to 3.3 g oil feed/hr/g catalyst. They reported the trend of increasing desulfurization and denitrogenation with increasing space time. However, no indication was made with respect to the influence of pressure on desulfurization and denitrogenation in this continuous process.

A comparison of hydrocracking data is difficult to make. Hydrocracking includes reactions such as the breaking down of larger molecules to smaller ones and addition of hydrogen to molecules by saturation, replacement, etc. In this work, hydrocracking is considered as converting high boiling components of the coal-derived liquid to lower boiling components. Since a variety of feed oils and bases of measuring the extent of hydrocracking were used by previous workers, comparison of their works is difficult. However, reports on hydrocracking are available in numerous sources (4, 9, 10, 15, 16, 24). No attempt was made to correlate across the many feedstocks which have been

reported.

Since coal liquids contain a broad variety of hydrocarbon components, it would be extremely difficult to determine the kinetics and mechanism of the reactions involved with each of the components. Most research workers try to estimate the kinetics and mechanism of hydrocracking from product distribution data. Qader and Hill (16) reported that the mechanism of hydrocracking of a low temperature tar involves simultaneous and consecutive cracking, hydrogenation, and isomerization reactions. Removal of sulfur and nitrogen is caused by rupture of carbon-sulfur and carbon-nitrogen bonds followed by hydrogenation reactions (4, 11, 16).

In 1968, Qader, et al. (17) reported that the hydrocracking reactions of low temperature tar are all first-order with respect to gasoline formation, desulfurization, denitrogenation, and deoxygenation in the temperature range 752°F (400°C) to 932°F (500°C) at 1500 psig. Later, Qader, et al. (17) reported that the formation of gasoline obeys first-order kinetics in the pressure range of 1500 to 2500 psig, but the overall reaction order for gasoline formation from low temperature tar becomes two under low hydrogen pressure from below 1500 to 500 psig in a temperature range of 752°F (400°C) to 887°F (475°C). Hill, et al. (4), using coal tar boiling from 356°F (180°C) to 653°F (345°C), also came out with a first-order kinetic for gasoline formation at 3000 psig pressure and at temperature range 662°F (350°C) to 887°F (475°C).

The recent literature indicates the following trends:

1. Desulfurization and denitrogenation of coal liquids have been shown to rise with increasing pressure and temperature in batch processing. Most of the work is reported in a temperature range of 662°F

(350°C) to 932°F (500°C) and a pressure range of 1000 psig to 3000 psig.

2. Desulfurization and denitrogenation of coal liquids have been shown to rise with increasing temperature and space time in continuous processing with the temperature range of 640°F (338°C) to 790°F (421°C) and the pressure at 3000 psig.

3. Nothing has been found to study pressure as a variable of desulfurization and denitrogenation of coal liquids in continuous processing.

4. Some kinetic data from batch processing were reported, but none from continuous processing has been found.

5. Desulfurization, denitrogenation, deoxygenation, and gasoline formation were reported to obey first-order kinetics in the temperature range 752°F (400°C) to 932°F (500°C) at 1500 psig. The kinetics of gasoline formation was shown to follow first-order reaction at pressures from above 1500 psig to 3000 psig, and at temperatures from 662°F (350°C) to 932°F (500°C). However, the kinetics of gasoline formation was said to indicate an overall reaction order of two at pressures from below 1500 to 500 psig in the same temperature range.

CHAPTER III

EXPERIMENTAL

A. Description of Equipment

The experimental system was constructed as shown in Figures 1 and 2 (see Table I for complete listing of the equipment).

The reaction system was a packed-bed reactor made from a ½ in O.D. stainless steel tube packed with inerts and catalyst. A thermowell ran through the central axis of the reactor, enabling the temperature at any point on the central axis of the reactor to be measured by a thermocouple which was connected to a digital temperature read-out. A heated tube line carrying liquid feed and another tube line carrying hydrogen gas met at the top of the reactor before entering the catalyst bed. The reactor was surrounded by heaters which were made up of square aluminum blocks grooved to hold beaded resistance heating wires. These are shown in Figure 3. The heating unit has three sections. The sections on the ends were controlled manually by variacs, and the middle section was controlled by a temperature programmer. The product that left the reactor could enter either a waste or a product receiver as controlled by the valving arrangement. In either receiver, the entering fluids travelled through a tube to the bottom, where the liquid disengaged and the gas bubbled through the liquid. The gas rose out of the receiver and through the back-pressure regulator which maintained

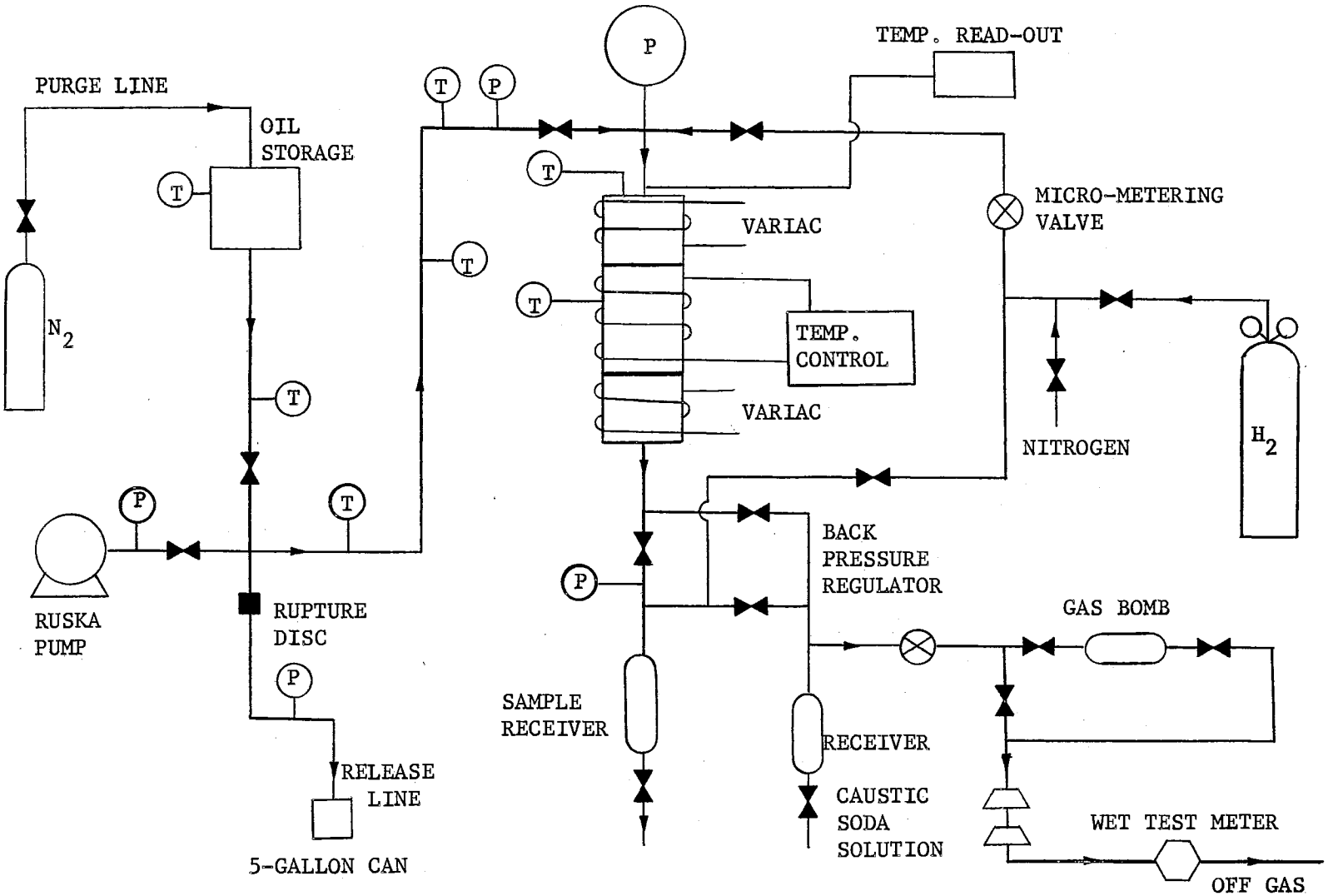


Figure 1. Experimental System.

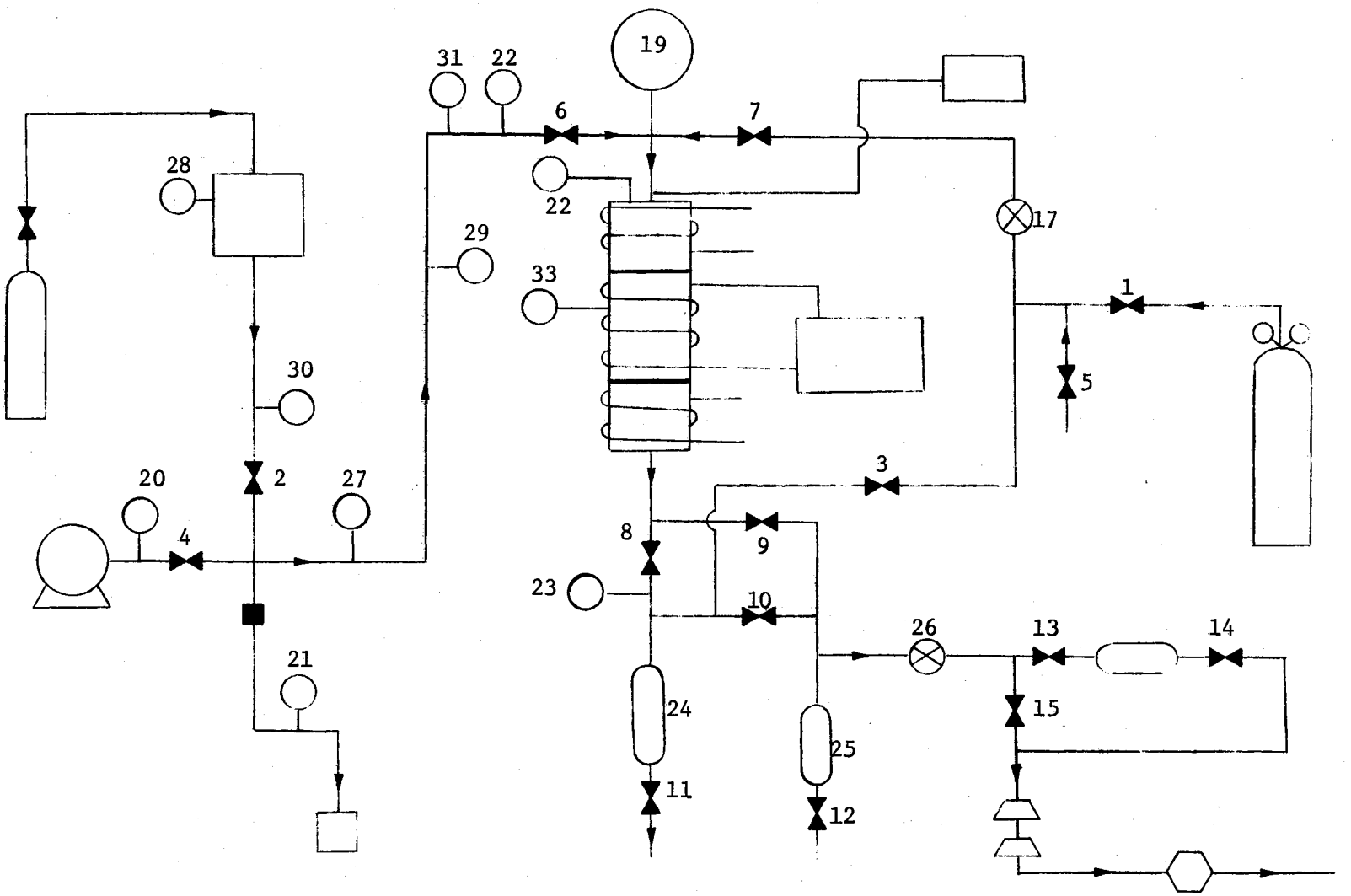
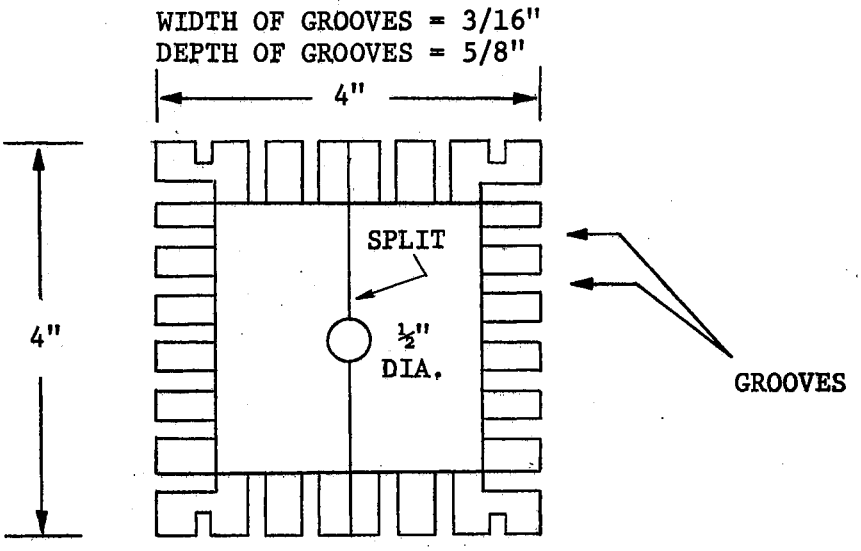
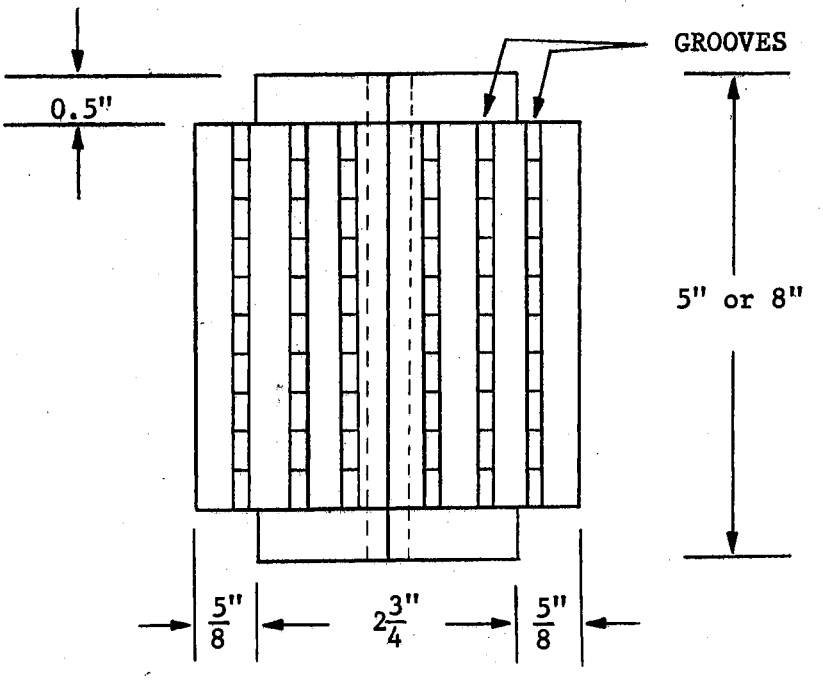


Figure 2. Experimental System, Numbered.



(a) Top View of Heating Block



(b) Side View of Heating Block

Figure 3. Aluminum Heating Block.

TABLE I

LIST OF EXPERIMENTAL EQUIPMENT

Tubing--for carrying liquid or gas, or both, ¼ in O.D., stainless steel.

Tubing--for carrying gas, 1/8 in O.D., stainless steel.

Reactor--½ in O.D., 0.049 in wall thickness, 316 stainless steel.

Valves 1, 3, 4, 5, 6, 7, 10, 12, 13, 14, and 15--¼ in, gate valves, 316 stainless steel, 9000 psia maximum, Autoclave 6V71UL4.

Valves 2, 8, 9, 11--½ in, gate valves, 316 stainless steel, Autoclave 6V71UL8.

Valve 17--¼ in, micro-metering valve, 316 stainless steel, Whitey 22RS4.

Pump--Ruska positive displacement pump, Model No. 2242 BI STQ, 500 cc bbl capacity, feed rates 2 to 240 cc/hr, max. pressure, 4000 psia.

Temp. Controller-- F&M Scientific 240 temperature programmer, automatic matching of output power, max. temperature 2000°F.

Pressure Gauge 19--Heise Bourdon Tube Gauge, 300 psig max. (calibration with Budenberg gauge showed max. deviation 3 psi less).

Pressure Gauge 20, 21, 22, and 23--Crosby Pressure Gauge, 300 psig max.

Receiver 24--316 stainless steel, 2250 ml, Matheson.

Receiver 25--316 stainless steel, 150 ml, Matheson.

Back Pressure Regulator--APCO, Model 1A, inlet pressure 400-3000 psig.

Digital Temperature Indicator--Doric Scientific DS-300-T3, temperature range 0-1595°F.

Feed Tank--8½ in O.D. and 7½ in high, stainless steel tank.

Wet Test Meter--Precision Scientific, 0.1 cu ft per revolution.

Gas Sample Bomb--stainless steel, 350 ml, Matheson.

Insulation material--fibre glass, Mcmaster Carr.

Thermocouple 27, 28, 29, 30, 31 and 32*--iron-constantan, 0.04 in O.D. type 304 stainless steel sheath, bare sensor tip configuration, ½ in, Conax

Thermocouple 33--chromel-alumel, 0.04 in O.D., type 304, stainless steel sheath, grounded sensor tip configuration, ½ in, Conax

*Thermocouple 32 calibrated vs. standard thermocouple in temp. range 550°F to 850°F.

high pressure inside the system. The gas passed on through the sample bomb, caustic soda solution, and wet test meter. There were valves at the ends of the sample bomb where a gas sample could be trapped inside. The gas could also by-pass the sample bomb, going directly through caustic soda solution, where hydrogen sulfide was removed. The wet test meter served to measure the flow rate of the off gas which was removed through a ventilating hood.

The liquid feed was stored in a tank where it was warmed and stirred before it was drawn into the system through a control valve by a Ruska pump. This same pump was also used to meter and feed liquid through the tube lines into the reactor at a constant flow rate. The tube line leading from the pump to the reactor was wound with heat tapes, which were controlled by variacs. Five thermocouples were located along this tube line.

At the bottom part of this tube line, there was a vent release system. A rupture disc was installed so that the fluid inside the system would break through the disc into a five-gallon waste can whenever the system pressure exceeded a certain level. In these experiments, 3000 psig rupture discs were used. A pressure gauge was installed ahead of the rupture disc as a prewarning of excess pressurization.

Facilities for passing nitrogen, oxygen, and hydrogen sulfide were also available. Gases were taken from commercial bottles on a once-through basis. An excess flow valve was placed in the hydrogen feed line.

B. Activation of Catalyst

The catalyst was available in the form of cylindrical pellets with a diameter of 1/8 in. It was crushed and screened to sizes between mesh 8 and mesh 10. The reaction tube was filled with 15.8 g of catalyst at the middle section and with inert particles at both ends. The catalyst was activated through calcining and sulfiding processes.

In the calcining process, oxygen was allowed to flow at 0.3 normal cu ft/hr through the reactor. The reactor bed was heated from 72°F (room temperature) to 600°F in one and one-half hours and was maintained at 600°F for two hours. During this entire period of heating, oxygen passed through the reactor. Then heat sources were shut off and oxygen flow was stopped. However, nitrogen was allowed to flow through the reactor for purge and rapid cooling.

In the sulfiding process, hydrogen sulfide saturated the reactor throughout the period when the reactor was being heated for about two hours from room temperature to 700°F. The catalyst was maintained at 700°F for another two hours with hydrogen sulfide. Again heat sources were shut off and hydrogen sulfide was replaced by nitrogen purge for cooling. (Refer to Table II for list of chemicals; Table III for list of catalyst properties.)

C. Experimental Procedure

A pressure test using nitrogen was applied to the system before the start of each experiment. When no leakage was present, the feed oil (see Table IV for its properties) which was well stirred in the feed tank, was drawn into the system. Valve 2 was opened to allow the feed oil to be drawn into the barrel of the Ruska pump. Valve 6 was closed

TABLE II

LIST OF CHEMICALS USED*

Hydrogen--prepurified, 99.95%, 3500 psig, Matheson Company, Inc.

Nitrogen--purity 99.997%, 2200 psig, Matheson Company, Inc.

Hydrogen Sulfide--purity 99.6%, 252 psig, Matheson Company, Inc.

Oxygen--purity 99.5%, 2100 psig, Linde.

Inert Reaction Packing--¼ in, semiporcelain, berl saddle, Maurice A. Knight.

Caustic Soda Flakes--Chemistry Department

*Chemicals used in analyses are listed in Appendix A.

TABLE III

CATALYST PROPERTIES

Nalcomo 474

*CoO, wt%	3.5
*MoO_3, wt%	12.5
Support	Alumina
Pore Volume, cc/g	0.463
*Surface Area, m^2/g	270
Pellet Density, g/cc	1.31
*Packed Bed Density, g/cc	0.73
Pellet Size	8/10 Mesh

*Nalco Data.

TABLE IV

FEED OIL PROPERTIES

Carbon, wt%	90.65
Hydrogen	5.76
Sulfur	0.486
Nitrogen	0.905
Ash	Nil
API Gravity @ 60°F	-7

*Distillation

Initial	380°F	193.3°C
10 vol%	450	232
30	570	299
50	650	343
70	700	371
90	815	435

*Normal boiling data were estimated from ASTM D 1160 data taken at 50 mm Hg absolute.

to prevent fluid from the reactor or the gas supply line to be drawn into the oil storage line. Valve 2 was closed when the barrel was filled with oil. The reactor was pressurized by sending compressed hydrogen through the system by opening valves 1, 18, 7, 8, 10, and 15. The back pressure regulator was adjusted to maintain a desired pressure in the reactor. The reactor was brought up to reaction temperature under this hydrogen flow. The feed oil in the storage barrel of the Ruska pump was first heated and was then pressurized by the pump to the same pressure as that of the hydrogen in the reactor. When the desired temperature profile was reached, feed oil was then pumped into the reactor at a constant flow rate. Liquid samples could be collected in receiver 24 when the desired temperature profile and hydrogen flow rate were established. The amount of time required to collect a liquid sample depended upon the liquid flow rate controlled by the pump and the quantity of sample needed for analysis. Gas sample could also be collected in a gas bomb though the intention of this work was to study the liquid product only. Before taking a liquid sample, valves 8 and 10 had to be closed and valve 9 had to be opened at the same time. The sample was then collected into a bottle by opening valve 11 slowly. After sample collection, valve 11 had to be closed. Valve 17 was opened momentarily to bring the pressure in the sample receiver back to system pressure. Valves 8 and 10 had to be opened while valve 9 had to be shut simultaneously to start collecting the next sample.

The shut-down of an experiment included first shutting off the heat sources, liquid feed, and hydrogen feed, and also releasing the pressure of the system. The system was then purged with nitrogen, which helped cool the reactor and wash away any left-over oil from the

catalyst, thus preventing the formation of coke on the catalyst.

D. Sulfur Analysis

Sulfur contents of coal liquids were analyzed by means of a Leco automatic sulfur determinator which has a model 521-500 induction furnace and a model 532-000 automatic titrator. According to the Leco iodate method, approximately 2 ml of starch-potassium iodide (KI) solution were mixed with 80 ml of a 1% hydrochloric acid. When a small amount of potassium iodate (KIO_3) solution was introduced, iodine was formed according to the equation:

$$KIO_3 + 5KI + 6HCl \longrightarrow 3I_2 + 6KCl + 3H_2O$$

A complex formed by iodine and starch was blue in color:

$$I_2 + \text{starch} \longrightarrow \text{blue complex}$$

Azide was also added to hinder the acidic property of NO from influencing the titration step later. The reaction after combustion of sulfur followed the equation:

$$SO_2 + I_2 + 2H_2O \longrightarrow H_2SO_4 + 2HI$$

The volume of iodate solution required to maintain the original blue color against this bleaching action of SO_2 was an indication of the sulfur content in the sample. Such volume was read directly as percent sulfur in a Leco sulfur determinator. Details of the equipment and the operation procedure are available in the manual produced by Leco Laboratory Equipment Corporation, 1415 Hilltop Road, St. Joseph, Michigan, 49085. Few samples were degassed to drive off H_2S and NH_3 before analysis. However, no significant influence was observed.

In this work, a coal liquid was used as a calibration reference for the Leco furnace. The sulfur content of this reference mentioned

was obtained from Pittsburg and Midway Coal Mining Co. Briefly, the procedure calls for calibrating with this reference oil, and then using the calibration factor for routine analyses of experimental samples. A sample calculation illustrating the technique is presented in Appendix B.

E. Nitrogen Analysis

Nitrogen analysis is based on the method described by Kjeldahl (20). The analytical work consists of four steps, namely: digestion, neutralization, distillation, and titration.

1. Digestion

For hydrocarbon, the purpose of digestion in 1.84 specific gravity H_2SO_4 is to oxidize nitrogen to $(NH_4)_2SO_4$, carbon to CO_2, and hydrogen to H_2O. For the coal-derived liquid, the digestion is performed by heating a solution composed of one g of the coal-derived liquid, 10 g of Na_2SO_4, 10 g of $CuSO_4$, three large Hengar granules, and 25 ml of 1.84 specific gravity H_2SO_4.

2. Neutralization

The samples are neutralized to free NH_4^+ ions. The digested hydrocarbon is diluted with 200 ml of distilled water. It is then made basic with NaOH.

$$NH_4^+ + OH^- \longrightarrow NH_3 + HOH$$

3. Distillation

Distillation follows in order to drive NH_3 out. The basic solution is distilled immediately into a flask containing boric acid. NH_3 is trapped.

$$NH_3 + HBO_2 \longrightarrow NH_4^+ + BO_2^-$$

4. Titration

The distillate is titrated with H_2SO_4 to obtain a direct determination of nitrogen.

A sample calculation is given in Appendix B along with a list of chemicals used.

F. Product Distillation

The apparatus and procedure for distillation are well established in ASTM D 1160, and are not to be repeated here. The distillation curves are made by plotting temperature against volume. Such curves are used for calculation of percent conversion of hydrocracking as defined in Appendix B along with a sample calculation. The ASTM distillation was routinely made at 50 mm Hg. However, distillations were also attempted at 8 and 10 mm Hg, but pressure control proved to be more erratic at these lower pressures.

CHAPTER IV

EXPERIMENTAL RESULTS

The variables studied were temperature at 600°F (315.6°C), 700°F (371.1°C), and 800°F (426.7°C), pressure at 500, 1000, and 2000 psig, and space time from 0.216 to 1.802 cc catalyst/cc feed oil per hour. Criteria for measurement of results are defined below in terms of percent conversion.

Percent sulfur conversion or desulfurization

$$= \frac{\text{wt. of sulfur in feed} - \text{wt. of sulfur in liquid product}}{\text{wt. of sulfur in feed}} \times 100$$

Percent nitrogen conversion or denitrogenation is similarly defined.

Percent conversion of hydrocracking of 650°F + materials

$$= \frac{\left(\begin{array}{l}\text{vol. of liquid that boils}\\ \text{up to 650°F in product}\end{array}\right) - \left(\begin{array}{l}\text{vol. of liquid that boils}\\ \text{up to 650°F in feed}\end{array}\right)}{\text{vol. of liquid that boils above 650°F in feed}} \times 100$$

The percent conversion by hydrocracking of 500°F + materials is as above, except 650°F (343.3°C) is replaced by 500°F (260°C). The equivalent definitions used for calculation of results from distillation at 50 mm Hg are in Appendix B. The above definitions have not taken into account liquid expansion and gasification upon reaction.

The pressure, space time, and temperature effects on desulfurization are shown in Figures 4, 5, and 6. The uppermost limit of desulfurization within the boundary of the experimental conditions is 92 percent. Temperature has a distinct effect on desulfurization, since the

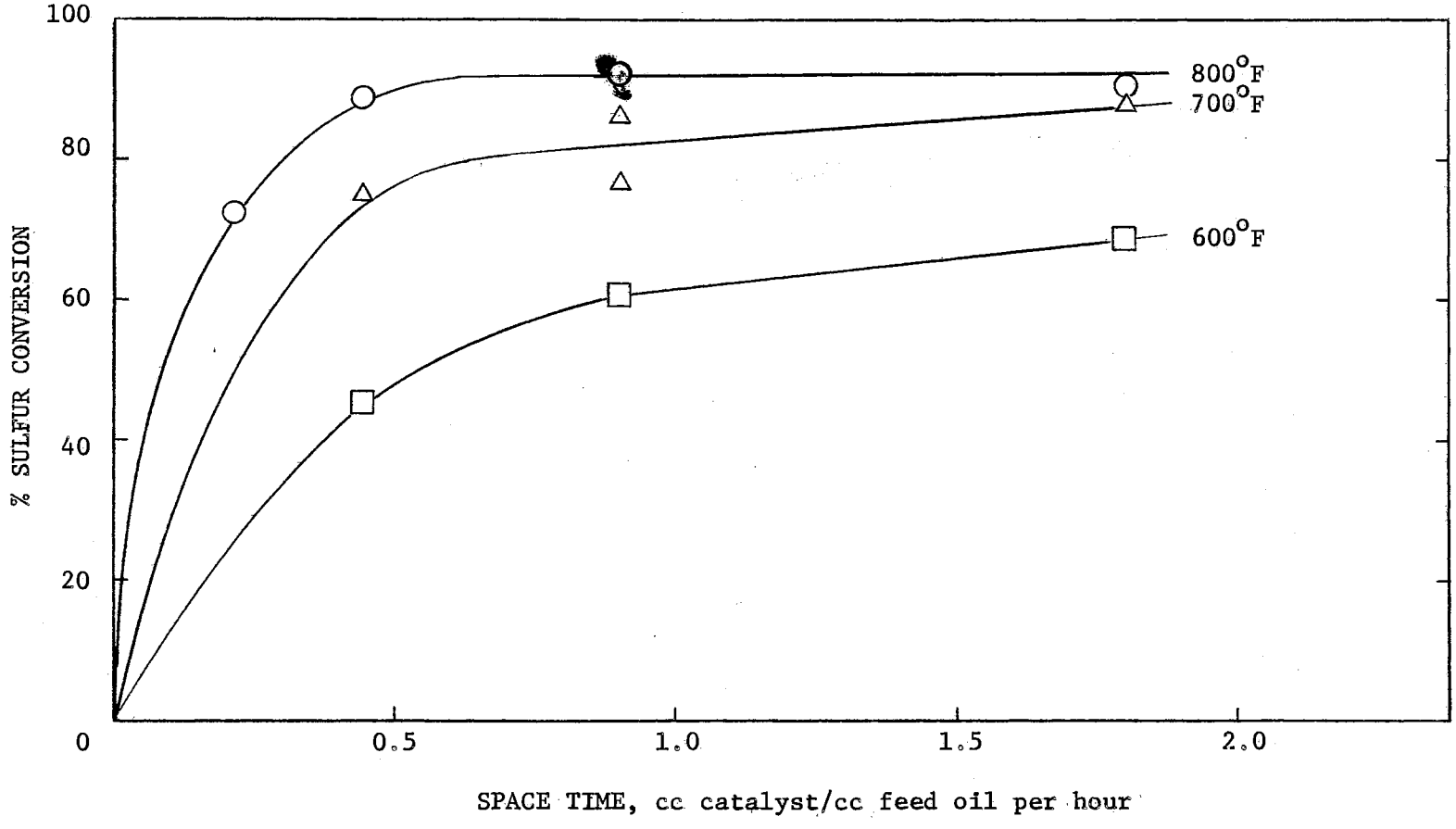


Figure 4. Desulfurization at 500 psig.

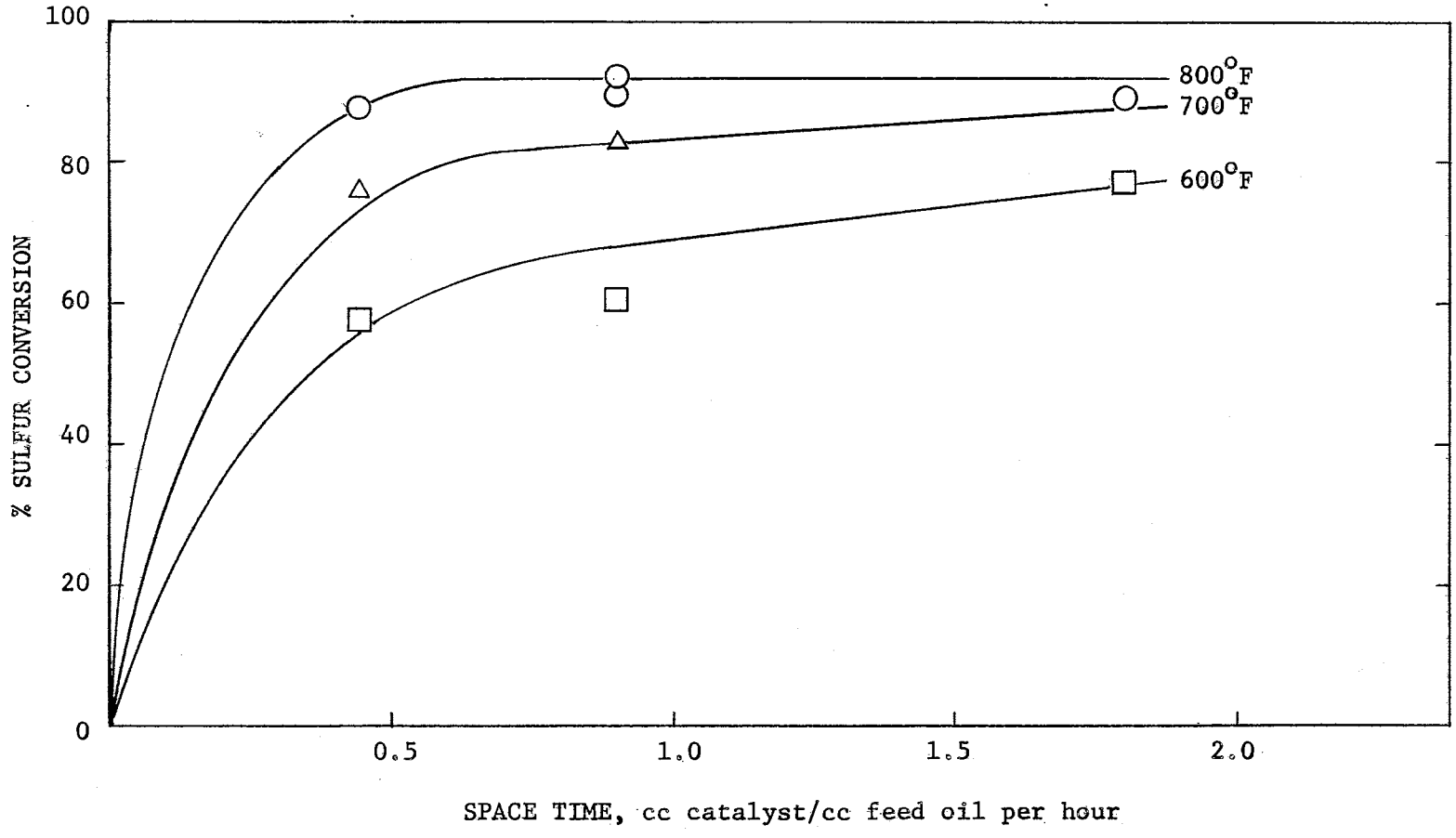


Figure 5. Desulfurization at 1000 psig.

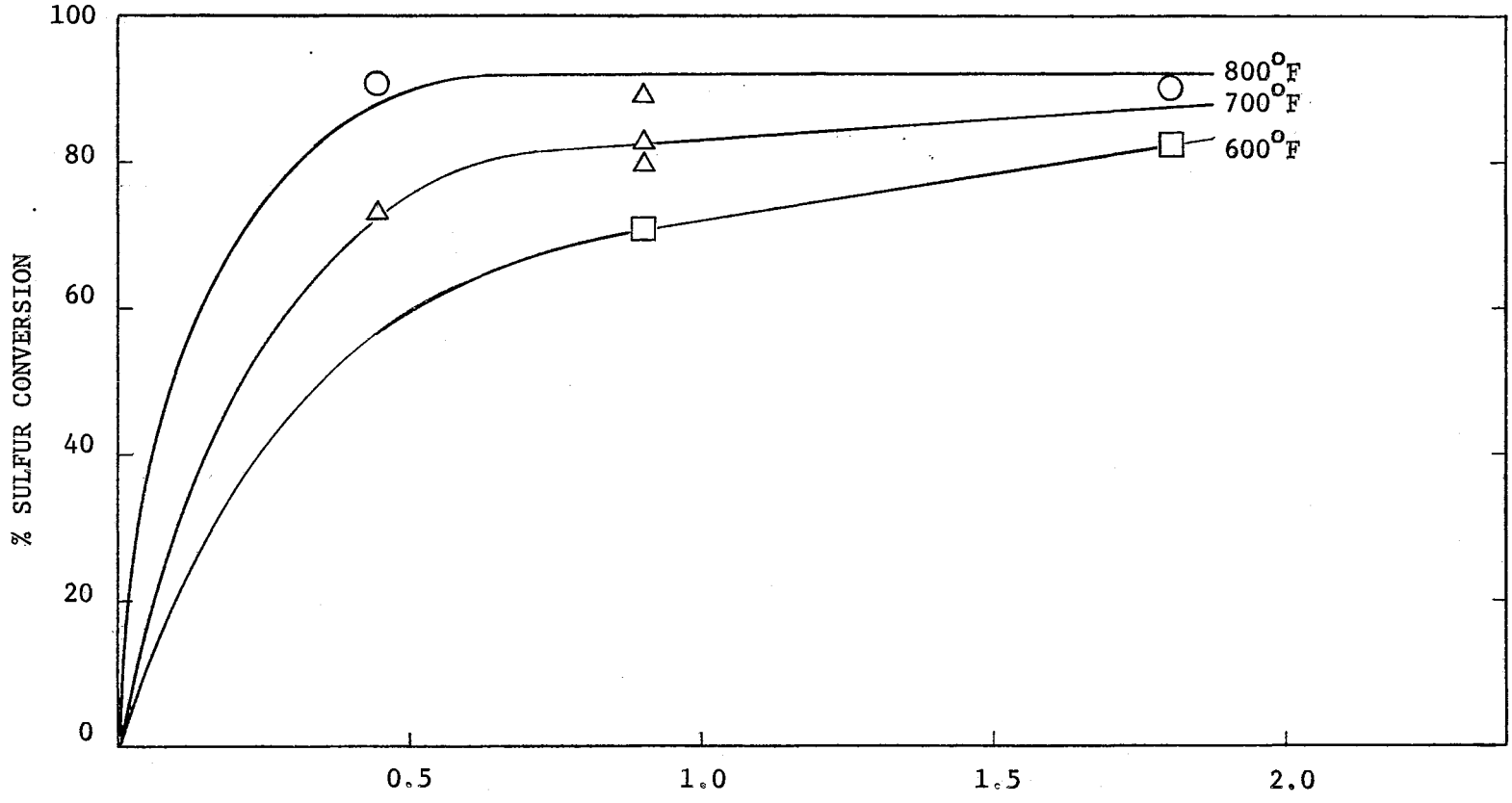


Figure 6. Desulfurization at 2000 psig.

percentage of sulfur removal increases with increasing temperature. The percentage of sulfur removal increases quite rapidly with increasing space time (based on the reciprocal of liquid velocity per volume of catalyst) up to 0.435 hr. At temperatures of 700° and 800°F, the rate of percent sulfur removal with respect to space time diminishes from space times 0.435 hr to 0.901 hr, and it is practically zero beyond space time 0.901 hr. At 600°F, the rate diminishes from space times 0.435 hr to 1.802 hr, which is the uppermost boundary of the experimental conditions for space time. Pressure has no noticeable effect on desulfurization at 700° and 800°F. However, at 600°F, pressure sensitivity is noticed as desulfurization increases with pressures from 500 psig to 2000 psig.

The duplicate points of 700°F and 0.901 hr space time on Figure 6 represent the sulfur removal results of two experiments which have the same conditions except that the oil-catalyst contacting times differed by about 60 hours. The oil-catalyst contacting time here is defined as the total length of time during which the catalyst has been contacting the oil at 600°F and above. There is not any detectable loss of desulfurization activity on the part of the catalyst for the series of experiments amounting to 70 hours of total oil-catalyst contacting time.

The effects of temperature, pressure, and space time on hydrocracking are shown in Figures 7 and 8, and Table V. The conversion of 650°F+ materials of the feed oil to lower boilers rises with increasing temperature. The highest conversion of 650°F+ materials to lower boilers in this sequence of experiments is 57 percent. The extent of influence that pressure and space time have upon the percent hydrocracking conversion is very much dependent on the level of temperature. At

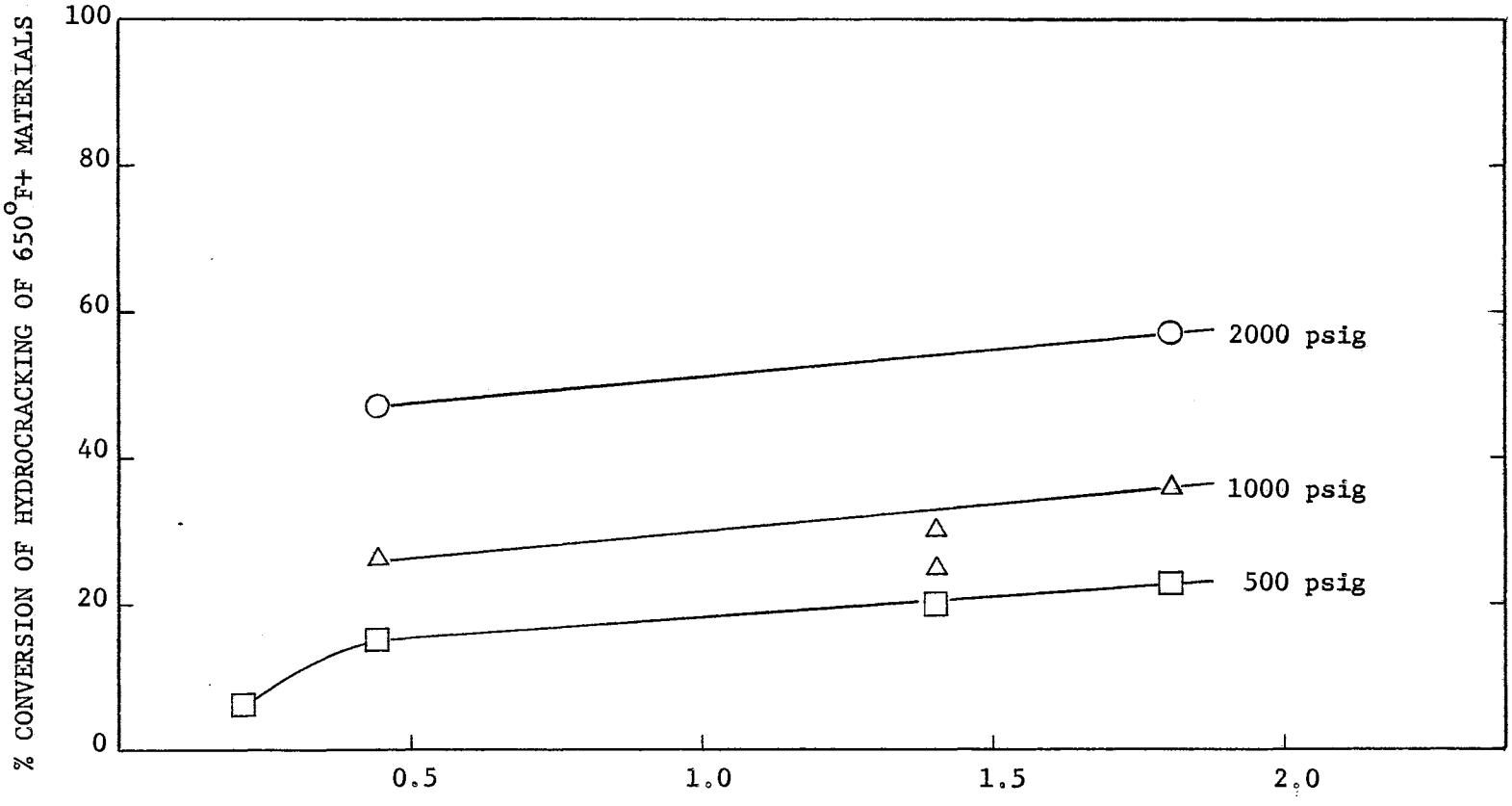


Figure 7. Hydrocracking at 800°F.

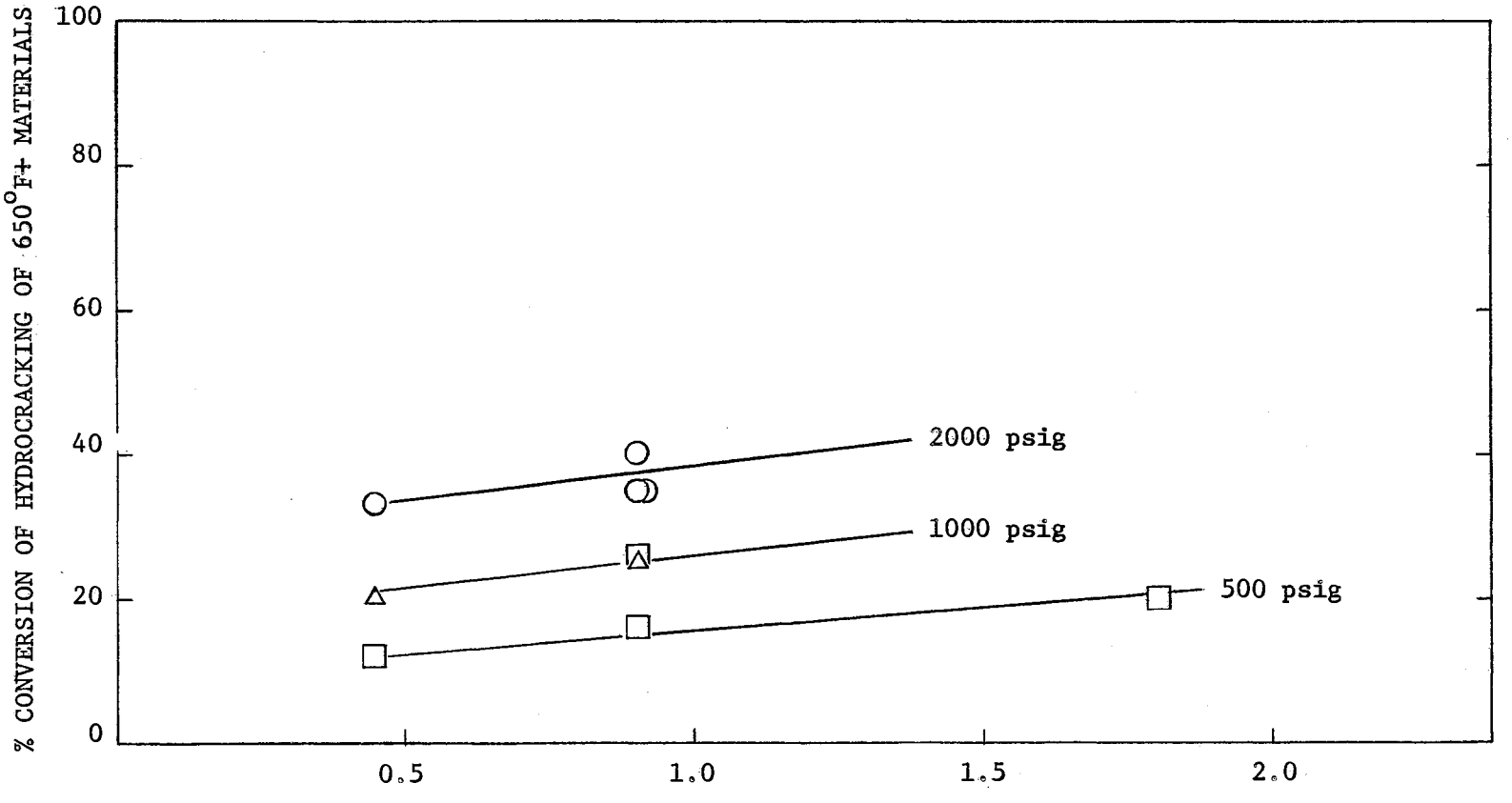


Figure 8. Hydrocracking at 700°F.

700°F and 800°F, the percent of 650°F+ materials converted to lower boilers rises with increasing space time, and it also rises with increasing pressure. However, at 600°F, the percent conversions are so low that the effects of space time and pressure are not outstanding. Maximum conversion of 650°F+ materials to lower boilers in the sequence of experiments operated at 600°F is 20 percent. In order to clarify the bases used for percent hydrocracking conversion here, it is necessary to point out that 17 vol. percent of the feed oil boils over at 500°F, and 50 vol. percent boils over at 650°F. Because of the low conversion at 600°F, to distinguish the results from a variety of pressures and space times with the analytical work developed thus far is very difficult. The conversions of 500°F+ materials to lower boilers are so low that it is not possible to suggest any trend for the effects of temperature, pressure, and space time. The percents conversion of 500°F+ materials with respect to temperature, pressure, and space time are shown in Table VI.

TABLE V

PERCENT CONVERSION OF HYDROCRACKING OF 650°F+ MATERIALS

	Pressure, psig		
	500	1000	2000
At 600°F			
*Space Time			
0.435	0	8	-
0.901	8	8	16
1.802	12	20	14

*Space times have units of cc catalyst/cc feed oil per hour.

TABLE VI

PERCENT CONVERSION OF HYDROCRACKING OF 500°F+ MATERIALS

	Pressure, psig		
	500	1000	2000
At 800°F			
*Space Time			
0.216	2.6	-	-
0.435	4.2	9.6	18.6
0.901	2.4	7.2, 7.2	-
1.802	6.0	9.6	21.0
At 700°F			
*Space Time			
0.435	1.2	2.7	8.4
0.901	0.6, 3.6	2.4	7.8, 3.6, 7.8
1.802	2.6	-	-
At 600°F			
*Space Time			
0.435	0	3.0	-
0.901	0	1.8	4.2
1.802	2.5	1.8	1.2

*Space times have units of cc catalyst/cc feed oil per hour.

The results on denitrogenation with respect to temperature, pressure, and space time are presented in Table VII. From Table VII, it is seen that denitrogenation data scatter badly. There is apparent inconsistency with respect to the level of denitrogenation at certain temperatures, pressures, and space times. The scatter of the data is caused by the analytical problems in determining nitrogen contents of product samples. Maximum denitrogenation achieved is 84.4 percent at 800°F, 2000 psig, and 1.802 cc catalyst/cc feed oil per hour. Minimum

denitrogenation is 5.6 percent at 600°F at 1000 psig, and 0.435 cc catalyst/cc feed oil per hour.

TABLE VII

PERCENT NITROGEN REMOVAL

	Temperature, °F		
	600	700	800
At 500 psig			
*Space Time			
0.216	-	-	19.1
0.435	26.3	32.4	38.3
0.901	20.0	59.1, 37.2	40.6
1.802	20.8	22.7	44.6
At 1000 psig			
*Space Time			
0.435	5.6	33.3	31.9
0.901	70.7	44.9	69.7, 61.2
1.802	19.4	-	69.6
At 2000 psig			
*Space Time			
0.435	-	42.3	74.8
0.901	5.7	55.9, 52.3, 56.9	-
1.802	12.7	-	84.4

*Space times have units of cc catalyst/cc feed oil per hour.

CHAPTER V

DISCUSSION

A heterogeneous process is most complicated when catalytic reactions on a solid surface involve interaction of reactants and formation of products in both liquid and gaseous phases (7). The gas-liquid-particle process in this work falls in this category. The process can be briefly described by the following elementary steps:

1. Transfer of gaseous molecules from the bulk gaseous phase through the gas-liquid interface into the bulk liquid phase.

2. Transfer of reactants from the bulk liquid phase to the external and internal surfaces of the porous catalyst.

3. Adsorption of reactants before reaction and desorption of reactants after reaction.

4. Transfer of products from pores and outer surfaces of the catalyst into the bulk liquid phase.

5. Transfer of gaseous products from the bulk liquid phase through the gas-liquid interface into the bulk gaseous phase.

However, there are times when the above steps cannot truly represent the process, owing to non-ideal operability of the trickle-flow reactor. In trickle-flow operation, the liquid flows downward through a packed bed reactor, and the gas goes either cocurrent or countercurrent to the liquid. The liquid does not necessarily flow in a

continuous phase over the solid particles, as does the gas. Maldistribution of liquid owing to flow on reactor wall surface and channels may be a problem. Various studies have been made on liquid distribution (6, 18, 19, 23). Some workers related maldistribution to the ratio between reactor tube diameter and packing diameter. Different criteria have been set for this ratio above which maldistribution is believed to be insignificant. However, no one criterion is agreed upon. Weimann and Satterfield, et al. (18, 23) stated that a ratio of 25 to 1 is desirable to avoid maldistribution, although a ratio as low as 5 to 1 has also been recommended. On the other hand, De Wall and Van Mameren (6) stated that when the liquid is evenly distributed above the packing, it stays evenly distributed in the packing. In this work, the ratio is 6.2 to 1.

Another problem associated with trickle-flow operation is backmixing. The undesirable effect of backmixing on overall conversion and the mathematical description of such an effect assuming first-order reaction are available in standard text (1). In recent work, Mears (12) related the ratio between the height of catalyst packing and packing diameter to backmixing. For a first-order reaction, the ratio of 350 is a conservative estimate to ensure freedom from backmixing. The ratio in this work is 144, though the reaction order is not determined.

No attempt is made to determine the order of any reaction in this work. Studying reactions of low temperature tar, Qader, et al. (17) showed that desulfurization, deoxygenation, and denitrogenation are first-order reactions at a constant hydrogen pressure of 1500 psig and for a temperature range of 400°C (752°F) to 500°C (932°F).

Superficial gas velocity can affect operations in trickle beds.

These might include liquid distribution, vaporization of liquid, partial pressure of gaseous components, and turbulence at gas-liquid interface. As shown in Table IX, experiments CN11 and CN12 were designed to test the influence of superficial gas velocity. The gas flow rate in experiment CN12 is ten times that in experiment CN11. However, results from both experiments show no significant difference. Thus, superficial gas velocity in the range of 0.6 to 6.0 normal cu ft/hr is not an important variable. These flow rates are equivalent to 3980 and 39,800 normal cubic feet of hydrogen per barrel of oil.

Temperature control in trickle-flow operation has been known to be a problem. Poor temperature control might produce high-temperature spots, which in turn can cause discontinuous liquid phase and vapor filled pores, thus adding resistance to mass transfer. Nonisothermal profiles will cause difficulty in both analysis and reproducibility of the data. Difficulties were also encountered by earlier workers in keeping a constant temperature along their reactors. Anderson, et al. (14) operated a one-inch reactor and reported that the maximum and minimum temperatures of the bed were generally within $\pm$ 20°F of its average temperature. Hawk, et al. (9) used a one-inch reactor with resistant wire elements wound on it to serve as heaters. In one series of experiments, they intended to run them at 400°C, but the average temperatures of the experiments ranged from 399° to 412°C. The magnitude of fluctuation of any temperature profile was not mentioned. In scaling up the operation from a 3/4-inch reactor to a 4-inch reactor, Berkebile, et al. (2) experienced severe temperature control problems. As a result, optimum selectivity and heteroatom removal could not be maintained.

In view of the temperature-controlling difficulty in the earlier work, special attention was placed on the design of the heating system in this work. The heaters described in the experimental section consisted of grooved aluminum blocks wound with beaded heat wires. The excellent conductivity of aluminum narrowed the temperature range across the connected heaters, thus producing an almost flat temperature profile. A typical temperature profile in the reactor bed is shown in Figure 9. The heating system used here appears to be an improvement over those used in the past. In addition, the heaters can be opened and a new reactor quickly installed.

The feasibility of a catalyst for industrial purposes is partly dependent upon the active life of the catalyst. The active life of the catalyst is the period of time during which the catalyst is operating to yield above a specified removal or conversion level. The major purpose in this case is heteroatom removal. It is important for experiments of catalytic hydrogenation of coal liquids to be performed during the active life of the catalyst. However, reports on studies of hydrogenation of coal liquids very seldom mention the lives of the catalysts. In most cases, it was assumed that the experiments were performed during the active period of the catalyst. Here, run CN6 and run CN18 (see Tables IX and X, Appendix C) were designed to check the activity of the catalyst. The catalyst betrayed no loss of activity after 70 hours of oil-catalyst contact.

The general effects of temperature and space time on desulfurization and denitrogenation found in this work agree well with those reported in the literature (2, 14, 15). However, the influence of pressure on heteroatom removal in continuous process was often not

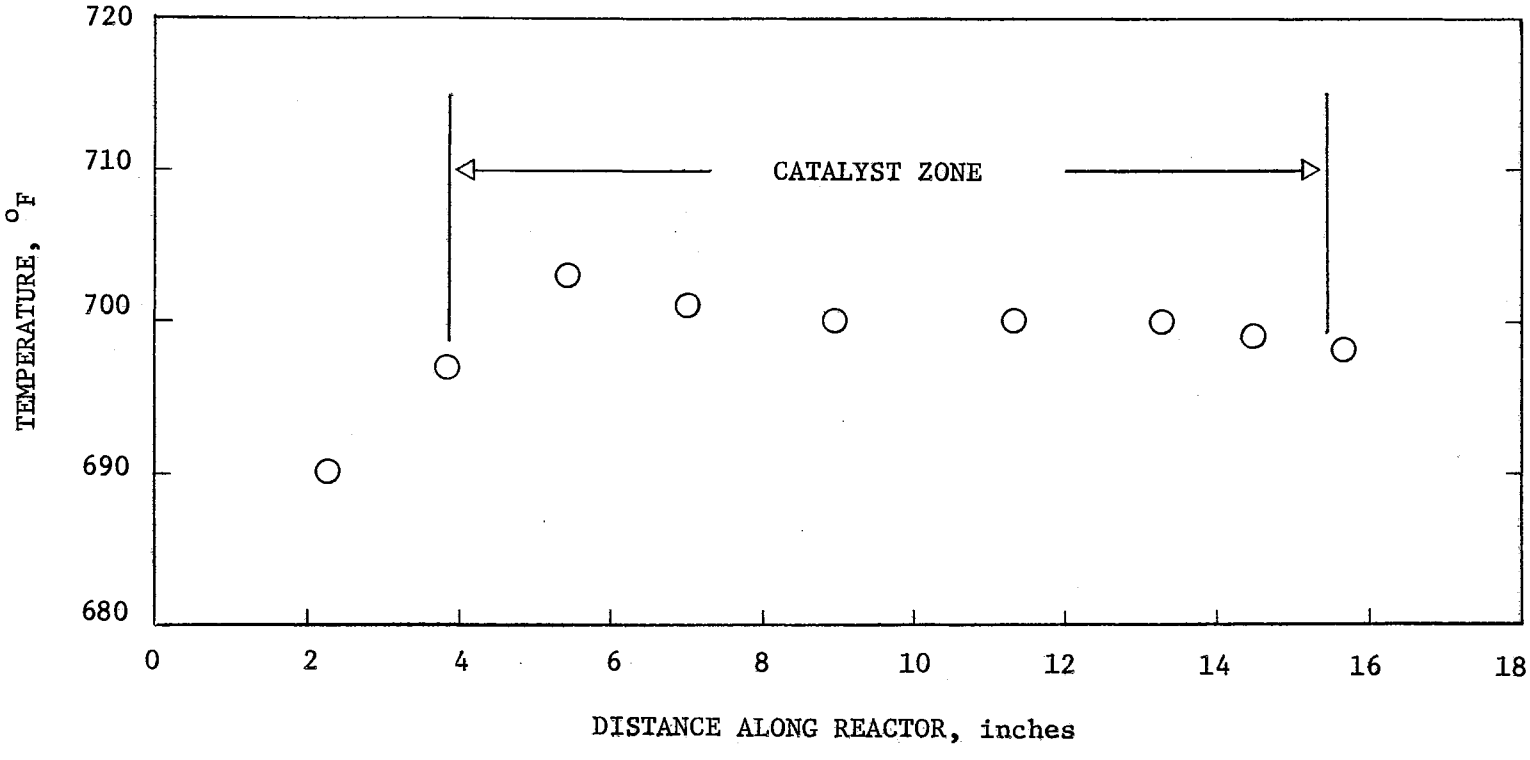


Figure 9. Typical Reactor Temperature Profile.

reported from studies concerning hydrogenation of coal liquids. From results of experiments done here (see Table VIII), the effect of pressure on desulfurization is not noticeable above 600°F, whereas at 600°F, desulfurization rises with increasing pressure. As a wide variety of feed oils and many different bases for dividing the distilled products were used by previous workers, comparison of hydrocracking is difficult. Nevertheless, the effects of pressure and temperature on hydrocracking agree well with King's observation (10). The proportion of low-boiling materials in the product increases with increasing pressure and temperature.

Sulfur content in the product was reduced to 400 ppm (see run CN12 in Table IX - Appendix C) which corresponds to 92 percent sulfur removal. Below this level (400 ppm), sulfur analysis is not reliable. Thus, 92 percent sulfur removal was the highest conversion detectable, even though conversions could have been greater than this. The desulfurization profiles in Figures 4, 5, and 6 all seem to level out at higher conversions (around 90 percent). This could be partly the analytical limitation, and not a rate limitation.

No experiments here were made below 600°F or above 800°F. This is because data obtained at 600°F indicate such low conversion, especially for nitrogen removal, that there is no incentive for further study at temperatures below 600°F. On the other hand, data obtained at temperatures in excess of 800°F have an excess degree of hydrocracking. For certain feed types, heteroatom removal should be achieved with only limited hydrocracking. Thus, no experiments were made beyond 800°F.

TABLE VIII

PERCENT SULFUR REMOVAL

	Pressure, psig		
	500	1000	2000
At 800°F			
*Space Time			
0.216	72.2	-	-
0.435	88.7	88.8	90.6
0.901	92.1	89.5, 92.1	-
1.802	90.6	88.9	90.0
At 700°F			
*Space Time			
0.435	74.9	75.7	73.0
0.901	76.5, 85.7	82.7	82.7, 79.5, 89.0
1.802	87.3	-	-
At 600°F			
*Space Time			
0.435	46.5	57.4	-
0.901	60.7	60.5	70.9
1.802	68.5	77.0	82.2

*Space times have units of cc catalyst/cc catalyst feed oil per hr.

Quality of the sample analysis and equipment performance depends a lot on the extent of their precision. For this reason, three groups of experiments (refer to Tables IX and X, Appendix C, for runs CN6, CN7, and CN18; CN9 and CN10; and CN11 and CN12), with each group having the same temperature, pressure, and space time, were designed to check the precision of the overall operation. The reproducibility of the samples from the overall operation is $\pm$ 0.044 wt. % for sulfur content (or $\pm$ 9.05 percent for desulfurization), $\pm$ 0.198 wt. % for nitrogen

content (or $\pm$ 21.2 percent for denitrogenation), $\pm$ 5.0 vol. % of the liquid at 500°F, and $\pm$ 2.5 vol. % of the liquid at 650°F. These numbers are an indication of the reproducibility in attempting to return to the same operating conditions with intermediate shut-down and start-up procedures. Since only one batch of catalyst was used, the variations of loading and pretreating new catalyst charges are not shown. The precision of the analytical work based upon 90 percent confidence limits is $\pm$ 0.009 wt. % for sulfur content, and $\pm$ 0.031 wt. % for nitrogen content. The D 1160 distillation method used was reported to have reproducibilities of 20 to 30 degrees F at 50 mm Hg absolute (21). (Refer to Tables IX, X, XI, and XII, Appendix C, for detailed experimental data.)

CHAPTER VI

CONCLUSIONS, RECOMMENDATIONS, AND CONSIDERATIONS

A. Conclusions

The following conclusions are made from this work:

1. Overall operability of the experimental system with respect to controlling of temperature, pressure, and feed rate, is satisfactory.

2. Experimental results reveal that desulfurization, denitrogenation, and hydrocracking increase with increasing temperature, pressure, and space time. Among the three variables, temperature has the most significant influence on desulfurization, denitrogenation, and hydrocracking.

3. The data collected here, except the denitrogenation data, can serve as a reference set for later catalyst development. At least the ranges of variables are established.

4. Reproducibility of denitrogenation data is unsatisfactory. This poor reproducibility is caused by inadequate analytical techniques.

5. This study is satisfactory as a preliminary step to develop apparatus and analytical techniques for a more detailed and refined program.

B. Recommendations and Considerations

The apparatus used in this work has all of the essential features to allow hydrogenation of the coal-derived liquid to take place at the

optimal conditions of pressure, temperature, and space time. However, modifications of the equipment can be adopted to make the operation more convenient and efficient. To avoid polluting the air inside the laboratory and to prevent inconvenient operation, another receiver should be connected below the sample receiver, so that liquid samples can be transferred safely from the sample receiver at system pressure to the lower receiver at approximately atmospheric pressure by operation on a valve between the two receivers. This way, the sample isolated at the lower receiver can be degassed to remove hydrogen disulfide and ammonia before it is exposed to the atmosphere for analysis. It will be interesting to estimate the amount of hydrocarbon vapor leaving with the off gas. Hydrocarbon content in the off gas can be analyzed by a gas chromatograph.

It is desirable to raise the ratio between the catalyst packing length and the catalyst size to avoid backmixing (12). The ratio required can be estimated from the studies done by Mears and Qader, et al. (12, 17). The ratio can be raised by either increasing the catalyst packing length, or decreasing the catalyst size. Of course, temperature control can become a problem depending on the catalyst packing length, and pressure drop can be a hindrance as to how small a packing diameter can be used. Though qualitative suggestions are made here, no specific study is attempted to evaluate the trade-off quantitatively.

Further studies can be made using this set of equipment, or with modifications, if desired. Firstly, studies can be made on pore size distribution of the catalyst carrier as a variable for heteroatom removal. Secondly, the effectiveness factor can be determined by

variation of particle size. Thirdly, comparison of different catalysts on heteroatom removal can be made. Fourthly, kinetic study on catalytic reactions of this system is possible.

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APPENDIX A

CHEMICALS USED FOR ANALYSES

A. Chemicals Used for Sulfur Analysis

1. LECO accelerators: iron powder and tin chips
2. MgO, magnesium oxide
3. KIO_3, potassium iodate
4. HCl, hydrochloric acid
5. Starch solution
6. NaN_3, sodium azide
7. Distilled water

B. Chemicals Used for Nitrogen Analysis

1. Na_2SO_4, sodium sulfate
2. $CuSO_4$, copper sulfate
3. H_2SO_4, sulfuric acid
4. NaOH, caustic soda
5. HBO_2, boric acid
6. Distilled water

APPENDIX B

SAMPLE CALCULATIONS FOR ANALYTICAL RESULTS

A. Calculation of Sulfur Content

$$\text{percent sulfur} = \frac{(\text{furnace factor})(\text{titration vol.} - \text{blank})}{\text{wt. of sample}}$$

$$\text{e.g.} \quad \%\ S = \frac{(490)(0.01950\ \text{ml} - 0.0003)}{107.9\ \text{g}}$$

$$= 0.0887 \text{ by wt.}$$

B. Calculation of Nitrogen Content

$$\text{percent nitrogen} = \frac{\left(\begin{matrix}\text{conversion for} \\ \text{chemical equivalence}\end{matrix}\right)\ (\text{vol. } H_2SO_4) \times 100}{\text{wt. of sample}}$$

$$\text{e.g.} \quad \%\ N = \frac{(0.175)(4.15\ \text{ml})}{1.0155\ \text{g}}$$

$$= 0.718 \text{ by wt.}$$

C. Readings From Distillation Curves

At 760 mm Hg, 17% by volume of the feed material boils over at 500°F (260°C), and 50% boils over at 650°F (343.3°C). At 50 mm Hg, 17% by volume boils over at 352°F (176.7°C), and 50% boils over at 465°F (240.5°C). Actual definitions of percent conversion by hydrocracking equivalent to those defined in Chapter IV but intended for results from distillation at 50 mm Hg are as the following:

Percent conversion by hydrocracking at 465°F+ materials

$$= \frac{\left(\begin{matrix}\text{volume of liquid that boils} \\ \text{up to } 465^{\circ}\text{F in products}\end{matrix}\right) - \left(\begin{matrix}\text{vol. of liquid that boils} \\ \text{up to } 465^{\circ}\text{F in feed}\end{matrix}\right)}{\text{vol. of liquid that boils above } 465^{\circ}\text{F in feed}} \times 100$$

The percent conversion by hydrocracking of 352°F+ materials is defined similarly as above,

e.g. % conversion of 650°F+ materials at 760 mm Hg

= conversion of 465°F+ materials at 50 mm Hg

$$= \frac{75\ \text{ml} - 50\ \text{ml}}{50\ \text{ml}} \times 100 = 50$$

APPENDIX C

EXPERIMENTAL DATA

TABLE IX

EXPERIMENTAL DATA

	Run No.						
	CN6	CN7	CN8	CN9	CN10	CN11	CN12
Temperature, ^{o}F	700	700	700	700	700	800	800
Pressure, psig	2000	2000	1000	500	500	1000	1000
H_2 Flow Rate, cu ft/hr	6	4.8	6	6	1.8	0.6	6
Space Time, cc cat/cc oil/hr	0.901	0.901	0.901	0.901	0.901	0.901	0.901
Hrs of Oil-Catalyst Contact	6.5	11.5	18	24	30	36	42
% Sulfur Removal	82.7	79.5	82.7	76.5	85.7	89.5	92.1
% Nitrogen Removal	55.9	52.3	44.9	59.1	37.2	69.7	61.2
% Conversion of $650^{o}F+$ Materials	40	35	25	16	26	30	25
% Conversion of $500^{o}F+$ Materials	7.8	3.6	2.4	0.6	3.6	7.2	7.2

TABLE X

EXPERIMENTAL DATA

	Run No.						
	CN13	CN14	CN15	CN16	CN17	CN18	CN19
Temperature, ^{o}F	600	700	600	800	800	700	700
Pressure, psig	500	500	500	500	500	2000	2000
H_2 Flow Rate, cu ft/hr	3	3	3	3	3	4.8	3
Space Time, cc cat/cc oil/hr	0.435	0.435	0.901	0.901	0.435	0.901	0.435
Hrs of Oil-Catalyst Contact	46	49	55	61	64.5	70	72.5
% Sulfur Removal	46.5	74.9	60.7	92.1	88.7	89.0	73.0
% Nitrogen Removal	26.3	32.4	20.0	40.6	38.3	56.9	42.3
% Conversion of $650^{o}F$+ Materials	0	12	8	20	15	35	33
% Conversion of $500^{o}F$+ Materials	0	1.2	0	2.4	4.2	7.8	8.4

TABLE XI

EXPERIMENTAL DATA

	Run No.						
	CN20	CN21	CN22	CN23	CN24	CN25	CN26
Temperature, ^{o}F	800	800	600	600	700	800	600
Pressure, psig	2000	1000	1000	1000	1000	500	500
H_2 Flow Rate, cu ft/hr	3	3	3	3	3	3	3
Space Time, cc cat/cc oil/hr	0.435	1.802	0.901	0.435	0.435	1.802	1.802
Hrs of Oil-Catalyst Contact	75.5	86.3	92.9	95.6	98.2	109.2	120.0
% Sulfur Removal	90.6	88.9	60.5	57.4	75.7	90.6	68.5
% Nitrogen Removal	74.8	69.6	70.7	5.6	33.3	44.6	20.8
% Conversion of $650^{o}F+$ Materials	47	36	8	8	20	22.6	20
% Conversion of $500^{o}F+$ Materials	18.6	9.6	1.8	3.0	2.7	6.0	2.5

TABLE XII

EXPERIMENTAL DATA

	Run No.						
	CN27	CN28	CN29	CN30	CN31	CN32	CN33
Temperature, ^{o}F	600	600	800	600	700	800	800
Pressure, psig	2000	2000	1000	1000	500	500	2000
H_2 Flow Rate, cu ft/hr	3	3	3	3	3	3	3
Space Time, cc cat/cc oil/hr	1.802	0.901	0.435	1.802	1.802	0.216	1.802
Hrs of Oil-Catalyst Contact	130.4	136.0	138.8	149.6	160.8	162.9	173.5
% Sulfur Removal	82.2	70.9	88.8	77.0	87.3	72.2	90.0
% Nitrogen Removal	12.7	5.7	31.9	19.4	22.7	19.1	84.4
% Conversion of $650^{o}F+$ Materials	14	16	26	20	20	6	57
% Conversion of $500^{o}F+$ Materials	1.2	4.2	9.6	1.8	2.6	2.6	21.0

VITA[2]

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